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(Dated: June 14, 2006)

I present a detailed derivation of wideband optical pulses interacting with a Raman transition in the kind of scheme currently used to generate the ultra broadband light fields needed to create ultrashort pulses. In contrast to the usual approach using separate field envelopes for the pump, Stokes, and anti-Stokes spectral lines, I use a *single* field envelope. This requires the inclusion of few-cycle corrections to the pulse propagation. The single-field model makes fewer approximations and is mathematically (and hence computationally) simpler, although it does require greater computational resources to implement. The single-field theory reduces to the traditional multi-field one using appropriate approximations.

This report should be read along with the paper Phys. Rev. **72**, *033804 (2005) "Wideband pulse propagation: single-field and multi-field approaches to Raman interactions" by P. Kinsler, G.H.C. New [1] for proper context. This document is primarily intended as a complete (as possible) record of the calculational steps that were necessarily abbreviated (or omitted) from that published work. It is an edited version of a longer document from which on-going work has been excised; and, as a "work in progress", despite my best efforts, may contain occasional mistakes. Please contact me if you have any comments, corrections or queries.*

[*] I worked at this institution while doing the bulk of this calculation. My main project was with Jon Marangos & Prof. GHC New on ultrabroadband multifrequency Raman generation, and I was funded with money from the EPSRC.

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I. INTRODUCTION

An important aim of current wideband Raman experiments is to try to efficiently generate few-cycle pulses [2, 3, 4, 5]. If driven strongly enough, the two-photon Raman transition modulates the incoming field by adding sidebands separated by the transition frequency. Wideband fields are generated as these sidebands generate sidebands of their own (and so on), thus generating a wide comb of frequency components separated by the transition frequency. If a scheme can be implemented that adjusts the phases of each component appropriately, then few- or single- cycle optical pulses can be obtained (see e.g. [3]). Standard theoretical treatments of this process split the field into fields components centred on the teeth of this comb. The approach has the advantage that the components can be modeled reasonably well with slowly varying envelopes, but of course it has the disadvantage of needing to keep track of a large number of components.

In experiments like those of Sali et.al. [5, 6], the Raman transition is driven near-resonantly by a pair of intense pump pulses about 100fs long; compared to the transition frequency of about 130THz, the spectra of each pump pulse (and hence the generated sidebands) are relatively narrow. This means that a multi-component model is still not unreasonable, even if numerical considerations might demand that the arrays used to store these spectra overlap in frequency space. However, if we were to move to shorter pump pulses, or to a single (much shorter) pump pulse with enough bandwidth to efficiently excite the transition, we would reach the regime where the "teeth" from the spectral comb significantly overlap. At this point, not only would we be forced to move from an SVEA (Slowly Varying Envelope Approximation) solution of the field propagation to a more accurate Generalized Few-cycle Envelope Approximation (GFEA) [7, 8], but the utility of multiple field components becomes questionable. In this regime it can be advantageous to treat the field as a single unit, rather than splitting it into pieces. Note that this approach still differs from solutions of Maxwell's equations such as FDTD (finite difference time domain)[9] or PSSD (pseudospectral spatial domain)[10], because our single-field is based on a second-order wave equation, and uses a convenient choice of carrier function to define a field envelope.

Following these considerations, we now derive a *single-field* model for Raman generation, and, apart from that notable detail, follow an analogous path to that of Hickman, Paisner, and Bischel (HPB) [11]. In the model, we find that the coupling constants retain an oscillatory behaviour at the transition frequency, and that it is this that impresses the sideband modulation on the propagating field. Since the field is not only wideband, but contains significant sideband components, we need to propagate this (no longer slowly varying) field envelope using the GFEA. The necessity of allowing for these can be demonstrated by converting the single-field model into a multi-field counterpart – without the envelope-gradient corrections, we will not get a correct multi-field model.

A. Summary of the theory and the numerical implementation

We model the wideband Raman generation process in the following way. We specify the field frequencies (ω_i) of interest, which are usually at integer spacings of the transition frequency (ω_A) from the main pump laser frequency (ω_0). Each of these field components is described usin a standard envelope theory (i.e. as $A_i(t)$) with a time-history, allowing us to simulate pulses as they travel through the Raman medium. The Raman medium is modelled as a

two-level atom using a (its) Bloch vector (u, v, w), and this Bloch vector is driven by each combination of spectrally adjacent field components ($\sim \sum A_i A_{-1}$). Each of the field components A_i is driven by the atomic polarization ($\sim v$) in combination with its pair of adjacent field components (i.e. A_{i-1}, A_{i+1}).

Each field component evolves as

$$\begin{aligned}\partial_z A_j &= \frac{\sigma\omega_j\alpha_{12}}{\epsilon_0 c_0}\left\{-\left[v' - \imath u'\right] A_{j+1}\exp\left(+\imath(k'_{j+1} - k'_j)z - \imath\Delta t\right) + \left[v' + \imath u'\right] A_{j-1}\exp\left(+\imath(k'_{j-1} - k'_j)z + \imath\Delta t\right)\right\} \\ &\quad - (k_j - k_0)\, A_j \end{aligned} \tag{1.1.1}$$

The transition evolves as ($\rho'_{12} \equiv (u', v')$)

$$\partial_t \rho'_{12} = \left(-\gamma_2 + \imath\Delta + 2\imath g' \sum_j A_j^* A_j\right)\rho'_{12} + 8\imath f' \sum_j A_j A_{j-1}^* . w . e^{+\imath(k_j - k_{j-1})z - \imath\Delta t} \tag{1.1.2}$$

$$\partial_t w = -\gamma_1 (w - w_i) + 4\imath f' \sum_j \left[A_j^* A_{j+1}\rho'_{12} . e^{+\imath(k_{j+1} - k_j)z - \imath\Delta t} - A_j^* A_{j-1}\rho'^*_{12} . e^{+\imath(k_{j-1} - k_j)z + \imath\Delta t}\right] \tag{1.1.3}$$

Here σ is the number density of the atoms or molecules; γ_1, γ_2 are the population and polarization decay rates for the transition; f' is the field-transition coupling constant; g' is the stark shift coefficient; ω_j, k_j are the frequencies and wavevectors for the field components, Δ gives a rotating frame for $\rho_{12} \to \rho'_{12}$

Additionally, a Cauchy dispersion is applied to the propagation of the field components.

Because each of the field components has a time-history, this translates to a spectral width. In typical cases with roughly nanosecond pulse lengths, the bandwidth of each component will be tiny compared to the transition frequency, so there will be large uneventful gaps in the total spectrum. In contrast, if the pulse lengths drop to (say) roughly 100 femtoseconds, the bandwidths of the field components will form a noticeable fraction of the total spectrum

B. A comment on Cauchy dispersion

As regards the mismatch terms in the Raman model, Geoff New has remarked (email, 20040121) that there's a key point about the γ_n's that is not made properly in most of the McDonald et al publications; since it is usually said rather enigmatically that the γ's are "parameterized" by γ_1. The point is that if one assumes a Cauchy-type law for the refractive index variation, all the gamma's are linked by a recurrence relation, and so you only need to specify one of them, from which all the others will follow. The point is made properly (to GN's knowledge) only in ref [12].

II. SINGLE-FIELD WIDEBAND RAMAN

Note: This single field derivation does mean some of the approximations as to the "slowness" of the field variation seem somewhat stringent. However, since a conversion to a multi-field model is possible, it would seem the field variation constraints are less stringent than they would first appear.

A. Coupled wavefunction equations

I start by considering the wave function ψ of a single molecule (e.g. H_2) and the electric field E, and write the time-dependent wave function by expanding it in terms of the eigenfunctions in the field-free (i.e. $E = 0$) case. This means I can get the expansion coefficients by solving for an effective Schröedinger equation that contains a two-photon Rabi frequency created by means of an interaction term based on a field-dependent dipole moment. I assume a dispersionless medium and write all equations in terms of position z and retarded times $t = t_{lab} - z/c$. Here I follow the method of HPB [11], but use only a single E field rather than multiple components. Note that HPB use *Gaussian* units, so there can appear to be inconsistencies when comparing my formulae (in S.I.) to theirs.

I denote the known molecular eigenfunctions of the unperturbed Hamiltonian H_0 as $|n\rangle$, and their corresponding energies $\hbar W_n$. I want to obtain the solution to

$$\begin{aligned}
(H_0 + V)\,\psi &= \imath\hbar\frac{\partial\psi}{\partial t} && (2.1.1)\\
\text{for} \qquad V &= -dE && (2.1.2)\\
\psi &= \sum_n c_n e^{-\imath W_n t}\,|n\rangle\,, && (2.1.3)
\end{aligned}$$

where d is the electronic dipole moment operator and the c_n are a set of complex probability amplitudes.

A standard derivation for the equations of motion of the c_i co-efficients proceeds as –

$$\begin{aligned}
\imath\hbar\frac{\partial}{\partial t}\sum_i c_i e^{-\imath W_i t}\,|i\rangle &= (H_0+V)\sum_j c_j e^{-\imath W_j t}\,|j\rangle && (2.1.4)\\
\imath\hbar\sum_i\left\{-\imath W_i c_i e^{-\imath W_i t} + e^{-\imath W_i t}\frac{\partial c_i}{\partial t}\right\}|i\rangle &= (H_0+V)\sum_j c_j e^{-\imath W_j t}\,|j\rangle && (2.1.5)\\
\imath\hbar\,\langle n|\sum_i\left\{-\imath W_i c_i e^{-\imath W_i t} + e^{-\imath W_i t}\frac{\partial c_i}{\partial t}\right\}|i\rangle &= \langle n|\,(H_0+V)\sum_j c_j e^{-\imath W_j t}\,|j\rangle && (2.1.6)\\
\imath\hbar\left\{-\imath W_n c_n e^{-\imath W_n t} + e^{-\imath W_n t}\frac{\partial c_n}{\partial t}\right\} &= \hbar c_n W_n e^{-\imath W_n t} + \sum_j c_j e^{-\imath W_j t}\,\langle n| - d.E\,|j\rangle && (2.1.7)\\
\imath\hbar\left\{-\imath W_n c_n + \frac{\partial c_n}{\partial t}\right\} &= \hbar W_n c_n - \sum_j c_j e^{-\imath W_j t + \imath W_n t}\,\langle n|\,d.E\,|j\rangle && (2.1.8)\\
\imath\hbar\frac{\partial c_n}{\partial t} &= -\sum_j c_j e^{-\imath(W_j - W_n)t}\,\langle n|\,d.E\,|j\rangle\,. && (2.1.9)
\end{aligned}$$

We now use perturbation theory, & $d_{nm} = \langle n|\,\hat{d}\,|n\rangle$, following two independent (but related) strands.

1. *CASE (i): Electric field*

This strand leaves the perturbing potential as a function of electric field E, and does not replace it with a carrier-envelope description. Although apparently the simplest strategy, it is generally impractical as it imposes constraints on the field and other model parameters that are too restrictive to be useful.

$$\begin{aligned}
\imath\hbar\frac{dc_n}{dt} &= -\sum_i c_i d_{ni} E\exp\left[-\imath\left(W_i - W_n\right)t\right] && (2.1.10)\\
\text{... assume } c_n, d_{ni} &\quad \text{vary only slowly, so I can integrate just the exponentials} && (2.1.11)\\
c_i &= -\frac{1}{\imath\hbar}\sum_j c_j d_{ij} E\frac{\exp\left[-\imath\left(W_j - W_i\right)t\right]}{-\imath\left(W_j - W_i\right)} && (2.1.12)\\
&= \frac{1}{\hbar}\sum_j c_j d_{ij} E\frac{\exp\left[-\imath\left(W_j - W_i\right)t\right]}{W_i - W_j} && (2.1.13)
\end{aligned}$$

Now substitute the c_i solution into the dc_n/dt equations, and introduce the shorthand notation $W_{ij} = W_i - W_j$,

$$\imath\hbar\frac{dc_n}{dt} = -\sum_i \left[\frac{1}{\hbar}\sum_j c_j d_{ij} E \frac{\exp\left[-\imath W_{ji} t\right]}{W_i - W_j}\right] d_{ni} E \exp\left[-\imath W_{in} t\right] \tag{2.1.14}$$

$$= -\frac{1}{\hbar}\sum_i \sum_j c_j d_{ij} E \frac{\exp\left[-\imath W_{ji} t\right]}{W_{ij}} d_{ni} E \exp\left[-\imath W_{in} t\right] \tag{2.1.15}$$

$$= -E^2 \sum_j \frac{1}{\hbar}\sum_i d_{ni} d_{ij} c_j \frac{\exp\left[-\imath W_{jn} t\right]}{W_i - W_j} \tag{2.1.16}$$

$$= -\sum_j c_j \alpha_{nj} E^2, \tag{2.1.17}$$

where

$$\alpha_{nj} = \frac{1}{\hbar}\exp\left[-\imath W_{jn} t\right] \sum_i \frac{d_{ni} d_{ij}}{W_i - W_j} \tag{2.1.18}$$

$$\text{UNITS:} \quad [\alpha_{nj}] \equiv J^{-1} s^{-1}.Cm.Cm.\left(s^{-1}\right)^{-1} = C^2 m^2 J^{-1} = \left(JV^{-1}\right)^2 m^2 J^{-1} = Jm^2 V^{-2} \tag{2.1.19}$$

$$\text{UNITS:} \quad \left[\frac{\alpha_{nj}}{\hbar} E^2\right] \equiv Jm^2 V^{-2}.J^{-1} s^{-1}.\left(Vm^{-1}\right)^2 = J.J^{-1} \times V^{-2}.V^2 \times m^2.m^{-2} \times s^{-1} = s^{-1} \tag{2.1.20}$$

Since we are only interested in the end states $j = 1, 2$, between which the Raman transition occurs, we only need calculate c_1, c_2; however we still retain the sum over all i intermediates states, as they affect the coupling between 1 and 2. The diagonal couplings $\{\alpha_{nj}, n = j\}$ are real; but the off-diagonal couplings $\{\alpha_{nj}, n \neq j\}$ undergo complex oscillations according to the difference in their energy levels.

Their *frequency dependence* is discussed after the following subsection; HPB's corresponding parameters, which *do not oscillate*, were assumed to be frequency independent.

2. CASE (ii): Electric Field Envelope

This strand replaces the electric field E with a carrier-envelope description, but, unlike HPB, I use only a single carrier-envelope component rather than a set indexed by some integer j. This is necessary, because in the previous strand I ended up with coupling constants α_{nj} with strong frequency dependences.

I introduce the envelope and carrier [13] for the field:

$$E = \left[Ae^{-\imath\omega_0 t} + A^* e^{+\imath\omega_0 t}\right] \tag{2.1.21}$$

$$\text{so that} \quad \imath\hbar\frac{dc_n}{dt} = -\sum_l c_l d_{nl} \exp\left[-\imath\left(W_l - W_n\right)t\right]\left[Ae^{-\imath\omega_0 t} + A^* e^{+\imath\omega_0 t}\right] \tag{2.1.22}$$

$$\text{now use } l \to i; \text{ and assume } \quad c_n, d_{ni} \text{ vary only slowly, so I can integrate just the exponentials} \tag{2.1.23}$$

$$c_i = -\frac{1}{\imath\hbar}\sum_j c_j d_{ij}\left[A\frac{\exp\left[-\imath\left(W_j - W_i + \omega_0\right)t\right]}{-\imath\left(W_j - W_i + \omega_0\right)} + A^*\frac{\exp\left[-\imath\left(W_j - W_i - \omega_0\right)t\right]}{-\imath\left(W_j - W_i - \omega_0\right)}\right] \tag{2.1.24}$$

$$= \frac{1}{\hbar}\sum_j c_j d_{ij}\left[A\frac{\exp\left[-\imath\left(W_j - W_i + \omega_0\right)t\right]}{W_i - W_j - \omega_0} + A^*\frac{\exp\left[-\imath\left(W_j - W_i - \omega_0\right)t\right]}{W_i - W_j + \omega_0}\right] \tag{2.1.25}$$

Note the swap of $(W_j - W_i)$ to $-(W_i - W_j)$ in the denominators. Now substitute the c_i solution into the dc_n/dt

equations (using $l \to j$), and introduce the shorthand notation $W_{ij} = W_i - W_j$,

$$\imath\hbar\frac{dc_n}{dt} = -\sum_i \left\{ \frac{1}{\hbar}\sum_j c_j d_{ij} \left[A\frac{\exp\left[-\imath\left(W_{ji}+\omega_0\right)t\right]}{W_{ij}-\omega_0} + A^*\frac{\exp\left[-\imath\left(W_{ji}-\omega_0\right)t\right]}{W_{ij}+\omega_0}\right]\right\} d_{ni}E\exp\left[-\imath W_{in}t\right] \tag{2.1.26}$$

$$= -\frac{1}{\hbar}\sum_i\sum_j c_j d_{ij}\left[A\frac{\exp\left[-\imath\left(W_{ji}+\omega_0\right)t\right]}{W_{ij}-\omega_0} + A^*\frac{\exp\left[-\imath\left(W_{ji}-\omega_0\right)t\right]}{W_{ij}+\omega_0}\right] d_{ni}\left[Ae^{-\imath\omega_0 t}+A^*e^{+\imath\omega_0 t}\right]\exp\left[-\imath W_{in}t\right] \tag{2.1.27}$$

$$= -\frac{1}{\hbar}\sum_i\sum_j c_j d_{ni}d_{ij}\left[A\frac{\exp\left[-\imath\left(W_{jn}+\omega_0\right)t\right]}{W_{ij}-\omega_0} + A^*\frac{\exp\left[-\imath\left(W_{jn}-\omega_0\right)t\right]}{W_{ij}+\omega_0}\right]\left[Ae^{-\imath\omega_0 t}+A^*e^{+\imath\omega_0 t}\right] \tag{2.1.28}$$

$$= -\frac{1}{\hbar}\sum_i\sum_j c_j d_{ni}d_{ij}\left\{ A^2\frac{\exp\left[-\imath\left(W_{jn}+2\omega_0\right)t\right]}{W_{ij}-\omega_0} + AA^*\frac{\exp\left[-\imath W_{jn}t\right]}{W_{ij}-\omega_0} + A^*A\frac{\exp\left[-\imath W_{jn}t\right]}{W_{ij}+\omega_0} + A^{*2}\frac{\exp\left[-\imath\left(W_{jn}-2\omega_0\right)t\right]}{W_{ij}+\omega_0}\right\} \tag{2.1.29}$$

$$\approx -\frac{1}{\hbar}\sum_i\sum_j c_j d_{ni}d_{ij}AA^*\exp\left[-\imath W_{jn}t\right]\left\{\frac{1}{W_{ij}-\omega_0}+\frac{1}{W_{ij}+\omega_0}\right\};\quad \text{by discarding the } 2\omega_0 \text{ terms;} \tag{2.1.30}$$

$$= -\frac{1}{\hbar}\sum_i\sum_j c_j d_{ni}d_{ij}AA^*\exp\left[-\imath W_{jn}t\right]\frac{W_{ij}+\omega_0 \quad + \quad W_{ij}-\omega_0}{\left(W_{ij}-\omega_0\right)\left(W_{ij}+\omega_0\right)} \tag{2.1.31}$$

$$= -\frac{1}{\hbar}\sum_i\sum_j c_j d_{ni}d_{ij}AA^*\exp\left[-\imath W_{jn}t\right]\frac{2W_{ij}}{W_{ij}^2-\omega_0^2} \tag{2.1.32}$$

$$= -AA^*\sum_j c_j\sum_i\frac{1}{\hbar}d_{ni}d_{ij}\exp\left[-\imath W_{jn}t\right]\frac{2W_{ij}}{W_{ij}^2-\omega_0^2} \tag{2.1.33}$$

$$= -\sum_j c_j\alpha'_{nj}\quad .2\left|A\right|^2 \tag{2.1.34}$$

where

$$\alpha'_{nj} = \frac{1}{\hbar}\exp\left[-\imath W_{jn}t\right]\sum_i d_{ni}d_{ij}\frac{W_{ij}}{W_{ij}^2-\omega_0^2} \qquad = \frac{1}{\hbar}\exp\left[+\imath W_{nj}t\right]\sum_i d_{ni}d_{ij}\frac{W_{ij}}{W_{ij}^2-\omega_0^2} \tag{2.1.35}$$

These redefined α'_{nj} parameters still oscillate, as they must because unlike in the HPB derivation, there is no frequency difference between field components to cancel with the Raman transition frequency. The coupling also varies with frequency, which is discussed next.

B. Raman coupling parameters: approximations

I now discuss two particular (and vital) approximations applied to the Raman coupling parameters.

First, note that (as in HPB), I will take the indices 1 and 2 to correspond to the two states involved in the (Raman) transition of interest; these will be the 0 and 1 vibrational (or perhaps rotational) levels of the electronic ground state. Indices 3 and above will correspond to (quoting HPB) "translational motion on higher electronic states".

Note: I can see that assigning these indices to higher electronic states will conveniently keep the energy separations for transitions to greater than that of the $1 \leftrightarrow 2$ *transitions, but it's not so clear to me why I can ignore all the higher vibrational (or rotational) states.*

Since I am interested only in the Raman transition, I specialise the above equations for the coefficients c_n, calculating c_1 and c_2 only, and assuming that the $d_{12} = \langle 1|\, d\,|\rangle$ dipole moment is zero. This means we will only be including transitions between indices 1 and 2 that *go via one of the higher states* $j \geq 3$, since we still allow $d_{1j},\ d_{2j} \neq 0\ \ \forall j \geq 3$. Further, I solve for the coefficients for the higher states in terms of c_1 and c_2, in an adiabatic approximation justified when c_1 and c_2 vary only slowly compared to the exponential terms.

Note: Separate from the oscillations that occur in my coupling parameters (but not in HPB), there is the issue of frequency dependence *which applies to both HPB and my parameters.*

For both HPB (their eqn.(15)), and my field-carrier based derivation (eqn.(2.1.35)), the presence of the field carrier in the denominator is helpful. Since it can reasonably be assumed to be much greater than the inter(Raman)-level

energy differences, the fractional terms will be correspondingly small, and so the α_{nj} (α'_{nj}) parameters can be assumed to be independent of frequency (or at least nearly so).

This is not in any way obviously (or even partially) true for the no-carrier E based (eqn.(2.1.18), α_{nj}) parameters, which depend only on the difference in Raman levels – at best you might perhaps hope that the denominators were approximate multiples of each other.

1. Near-zero difference

Here I assume that the forward and backward transitions (between levels 2 and 1) have nearly the same amplitude. It is not self-evidently true, but HPB must have made an equivalent assumption. In any case, since $\alpha'^{*}_{12} - \alpha'_{21}$ is the difference of similar terms, it will be smaller by at worst a factor of two $((1+\delta-1)/(1+\delta+1) \sim \delta/2)$

$$\alpha'^{*}_{12} - \alpha'_{21} = \frac{1}{\hbar}\exp\left[+\imath W_{21}t\right]\sum_i d^*_{1i}d^*_{i2}\frac{W_{i2}}{W^2_{i2}-\omega^2_0} - \frac{1}{\hbar}\exp\left[-\imath W_{12}t\right]\sum_i d_{2i}d_{i1}\frac{W_{i1}}{W^2_{i1}-\omega^2_0} \tag{2.2.1}$$

$$= \frac{1}{\hbar}\sum_i d^*_{1i}d^*_{i2}\left[\frac{W_{i2}}{W^2_{i2}-\omega^2_0} - \frac{W_{i1}}{W^2_{i1}-\omega^2_0}\right]\exp\left[+\imath W_{21}t\right] \tag{2.2.2}$$

$$\approx 0 \tag{2.2.3}$$

This approximation allows me to equate α'_{21} to α'^{*}_{12}, and hence change $-\alpha'_{21}c^*_1c_1 + \alpha'^{*}_{12}c_2c^*_2 \longrightarrow \alpha'_{21}\left(c^*_2c_2 - c^*_1c_1\right)$ in the ρ_{12} equation below. This is a vital step in getting to a simple form equivalent to the Bloch equations.

2. Sum is double

This follows from the above assumption (as per HPB) that the forward and backward transitions (between levels 2 and 1) have nearly the same amplitude. This approximation allows me to replace α'_{21} with α'_{12} in the w equation below, which simplifies my notation and makes the coupling term match that in the ρ_{12} equation, important in getting to true Bloch equations.

$$\alpha'_{12} + \alpha'^{*}_{21} = \frac{1}{\hbar}\exp\left[-\imath W_{21}t\right]\sum_i d_{1i}d_{i2}\frac{W_{i2}}{W^2_{i2}-\omega^2_0} + \frac{1}{\hbar}\exp\left[+\imath W_{12}t\right]\sum_i d^*_{2i}d^*_{i1}\frac{W_{i1}}{W^2_{i2}-\omega^2_0} \tag{2.2.4}$$

$$= \frac{1}{\hbar}\sum_i d_{1i}d_{i2}\left[\frac{W_{i2}}{W^2_{i2}-\omega^2_0} + \frac{W_{i1}}{W^2_{i2}-\omega^2_0}\right]\exp\left[-\imath W_{21}t\right] \tag{2.2.5}$$

$$\approx \bar{\alpha}_{12}e^{-\imath W_{21}t} \tag{2.2.6}$$

3. Sum is cosine

I do not reduce the exponential sum to a (trig) cosine function, for potential use later when matching and/or differencing exponentials. However, this sum is does not occur, so the $\bar{a}_{12}$ is never used.

$$\alpha'_{12} + \alpha'_{21} = \frac{1}{\hbar}\exp\left[-\imath W_{21}t\right]\sum_i d_{1i}d_{i2}\frac{W_{i2}}{W^2_{i2}-\omega^2_0} + \frac{1}{\hbar}\exp\left[-\imath W_{12}t\right]\sum_i d_{2i}d_{i1}\frac{W_{i1}}{W^2_{i2}-\omega^2_0} \tag{2.2.7}$$

$$= \frac{1}{\hbar}\sum_i d_{1i}d_{i2}\left[\frac{W_{i2}}{W^2_{i2}-\omega^2_0}\exp\left[-\imath W_{21}t\right] + \frac{W_{i1}}{W^2_{i2}-\omega^2_0}\exp\left[+\imath W_{21}t\right]\right] \tag{2.2.8}$$

$$\approx \bar{a}_{12}\left[e^{-\imath W_{21}t} + e^{+\imath W_{21}t}\right] \tag{2.2.9}$$

Here I calculate the Stark shift term

$$\alpha'_{11}-\alpha'^{*}_{22} = \frac{1}{\hbar}\exp\left[-\imath W_{11}t\right]\sum_i d_{1i}d_{i1}\frac{W_{i1}}{W_{i1}^2-\omega_0^2} - \frac{1}{\hbar}\exp\left[+\imath W_{22}t\right]\sum_i d^*_{2i}d^*_{i2}\frac{W_{i2}}{W_{i2}^2-\omega_0^2} \tag{2.2.10}$$

$$= \frac{1}{\hbar}\sum_i\left[d^*_{i1}d_{i1}\frac{W_{i1}}{W_{i1}^2-\omega_0^2} - d^*_{2i}d_{2i}\frac{W_{i2}}{W_{i2}^2-\omega_0^2}\right] \tag{2.2.11}$$

Simplify assuming $W_{21}\ll W_{i1},W_{i2}$, while assuming $W_{i1}/\left(W_{i1}^2-\omega_0^2\right)\sim 1$; or, more accurately, that

$$1 \gg \frac{W_{21}^n}{W_{i1}^n}\frac{W_{i1}^2}{W_{i1}^2-\omega_0^2} \qquad \text{for} \quad n = 1,\ 2, \tag{2.2.12}$$

and keeping only the terms first order in $W_{21}/W_{i1}, W_{21}/W_{i2}$, etc

$$\alpha'_{11}-\alpha'^{*}_{22} = \frac{1}{\hbar}\sum_i\left\{|d_{1i}|^2\frac{W_{i1}}{W_{i1}^2-\omega_0^2} - |d_{2i}|^2\frac{W_{i1}+W_{21}}{W_{i1}^2+2W_{i1}W_{21}+W_{21}^2-\omega_0^2}\right\} \tag{2.2.13}$$

$$= \frac{1}{\hbar}\sum_i\left\{|d_{1i}|^2\frac{W_{i1}}{W_{i1}^2-\omega_0^2} - |d_{2i}|^2\frac{W_{i1}+W_{21}}{\left(W_{i1}^2-\omega_0^2\right)}\frac{1}{\left[1+\left(2W_{21}/W_{i1}+W_{21}^2/W_{i1}^2\right)W_{i1}^2/\left(W_{i1}^2-\omega_0^2\right)\right]}\right\} \tag{2.2.14}$$

$$= \frac{1}{\hbar}\sum_i\left\{|d_{1i}|^2\frac{W_{i1}}{W_{i1}^2-\omega_0^2} - |d_{2i}|^2\frac{W_{i1}+W_{21}}{W_{i1}^2-\omega_0^2}\left[1-\left(2\frac{W_{21}}{W_{i1}}+\frac{W_{21}^2}{W_{i1}^2}\right)\frac{W_{i1}^2}{\left(W_{i1}^2-\omega_0^2\right)}+...\right]\right\} \tag{2.2.15}$$

$$\approx \frac{1}{\hbar}\sum_i\left\{|d_{1i}|^2\frac{W_{i1}}{W_{i1}^2-\omega_0^2} - |d_{2i}|^2\frac{W_{i1}+W_{21}}{W_{i1}^2-\omega_0^2}\left[1-2\frac{W_{21}}{W_{i1}}\frac{W_{i1}^2}{\left(W_{i1}^2-\omega_0^2\right)}\right]\right\} \tag{2.2.16}$$

$$\approx \frac{1}{\hbar}\sum_i\left\{\left[|d_{1i}|^2-|d_{2i}|^2-|d_{2i}|^2\frac{W_{21}}{W_{i1}}\right]\frac{W_{i1}}{W_{i1}^2-\omega_0^2} - \left[2|d_{2i}|^2\frac{W_{21}}{W_{i1}}\right]\frac{W_{i1}^3}{\left(W_{i1}^2-\omega_0^2\right)^2}\right\} \tag{2.2.17}$$

Ignoring the W_{21} transition frequency terms, this is just the difference in the energy shifts of the two levels with field intensity. This is a purely *real* quantity, with no imaginary part.

C. Two photon Bloch equations

From now on, I use $\mathfrak{I}$ (a script "I") to represent E^2 if following on from II A 1 and eqns.(2.1.17, 2.1.18); or to represent $2|A|^2$ if following on from II A 2 and eqns.(2.1.34, 2.1.35). Since it is the envelope-carrier description of the field E which is most useful (in II A 2), for most purposes $\mathfrak{I}=2|A|^2$ holds; remember that it is difficult to maintain the accuracy of the approximations relied on above for the E^2 picture (in II A 1). I also drop the prime on the α'_{nj} parameters used in the (Case Iii)) "$2|A|^2$" electric field envelope model.

Since we are only interested in c_1, c_2, and because we keep only stationary or slowly varying terms, we can write equations for c_1, c_2 as –

$$\imath\hbar\frac{dc_1}{dt} = -\alpha_{11}\mathfrak{I}c_1-\alpha_{12}\mathfrak{I}c_2 \tag{2.3.1}$$

$$\Longrightarrow \qquad \imath\hbar\frac{dc_1^*}{dt} = +\alpha^*_{11}\mathfrak{I}c_1^*+\alpha^*_{12}\mathfrak{I}c_2^* \tag{2.3.2}$$

$$\imath\hbar\frac{dc_2}{dt} = -\alpha_{21}\mathfrak{I}c_1-\alpha_{22}\mathfrak{I}c_2 \tag{2.3.3}$$

$$\Longrightarrow \qquad \imath\hbar\frac{dc_2^*}{dt} = +\alpha^*_{21}\mathfrak{I}c_1^*+\alpha^*_{22}\mathfrak{I}c_2^* \tag{2.3.4}$$

$$\text{In matrix form:} \qquad \frac{d}{dt}\begin{bmatrix}c_1\\c_2\end{bmatrix} = -\frac{1}{\imath\hbar}\begin{bmatrix}\alpha_{11}\mathfrak{I} & \alpha_{12}\mathfrak{I}\\ \alpha_{21}\mathfrak{I} & \alpha_{22}\mathfrak{I}\end{bmatrix}\begin{bmatrix}c_1\\c_2\end{bmatrix}, \tag{2.3.5}$$

D. Alternative formulations: Kien et.al. (KLKOHS), Hickman et.al. (HPB)

This interlude compares my matrix equation for c_1, c_2 to those from Kien et.al.[14] (KLKOHS) and Hickman et.al. [11] (HPB). This is useful as a fixed point of refernce beween the approaches, enabling quick conversions between the parameter variables.

$$\text{cf. (KLKOHS) eqn.(13):} \qquad \frac{d}{dt}\begin{bmatrix} c_a \\ c_b \end{bmatrix} = -\frac{1}{\imath\hbar}\begin{bmatrix} \hbar\Omega_{aa} & \hbar\Omega_{ab} \\ \hbar\Omega_{ba} & \hbar\Omega_{bb}-\delta \end{bmatrix}\begin{bmatrix} c_a \\ c_b \end{bmatrix}, \tag{2.4.1}$$

$$\text{where } B_q \text{ are KLKOHS's } E_q \text{ envelopes:} \qquad E = \frac{1}{2}\left[B_q e^{\imath\Xi} + B_q^* e^{-\imath\Xi}\right] \tag{2.4.2}$$

$$\hbar\Omega_{aa} = \frac{\hbar}{2}\sum_q a_q B_q B_q^* \tag{2.4.3}$$

$$\hbar\Omega_{ab} = \frac{\hbar}{2}\sum_q d_q B_q B_q^* \qquad = \hbar\Omega_{ba}^* \tag{2.4.4}$$

$$\hbar\Omega_{bb} = \frac{\hbar}{2}\sum_q b_q B_q B_{q+1}^* \tag{2.4.5}$$

$$\text{cf. (HPB) eqn.(13):} \qquad \frac{d}{dt}\begin{bmatrix} c_1 \\ c_2 \end{bmatrix} = -\frac{1}{\imath\hbar}\begin{bmatrix} -H_{11} & -H_{12} \\ -H_{21} & -H_{22} \end{bmatrix}\begin{bmatrix} c_1 \\ c_2 \end{bmatrix}, \tag{2.4.6}$$

$$\text{where } \alpha_{Hij} \text{ are HPB's } \alpha_{ij} \text{ parameters:} \qquad H_{11} = -\frac{1}{4}\sum_j \alpha_{H11}(\omega_j) V_j V_j^* \tag{2.4.7}$$

$$H_{12} = -\frac{1}{4}\sum_j \alpha_{H12}(\omega_j) V_j V_{j-1}^* \qquad = H_{21}^* \tag{2.4.8}$$

$$H_{22} = -\frac{1}{4}\sum_j \alpha_{H22}(\omega_j) V_j V_j^* \tag{2.4.9}$$

Thus comparing the Rabi-like parts of my eqn (2.3.5) with that of KLKOHS (my eqn (2.4.1)) and that of HPB (my eqn (2.4.6)), gives

$$\alpha_{12}\mathcal{I} = \alpha_{12}.2A^*A = \hbar f'.2A^*A \qquad = \frac{\hbar}{2}\sum_q d_q B_q B_{q+1}^* = \hbar\sum_q d_q.\frac{1}{2}B_q B_{q+1}^* \qquad = -\frac{1}{4}\sum_j \alpha_{H12}(\omega_j) V_j V_{j-1}^* \tag{2.4.10}$$

- This uses KLKOHS: $E = \frac{1}{2}\left(B_1 + B_1^* + B_2 + B_2^*\right)$
$\rightarrow E^2 = \frac{1}{4}\left(B_1^2 + B_1^{*2} + B_2^2 + B_2^{2*} + 2B_1B_1^* + 2B_2B_2^* + 2B_1B_2^* + 2B_1^*B_2\right)$
$\rightarrow E_{1-2}^2 \sim \frac{1}{4}2B_1B_2^* = \frac{1}{2}B_1B_2^*$
which tells us the size of the field contribution to the KLKOHS Rabi-like term.
- This uses HPB: which tells us the size of the field contribution to the HPB Rabi-like term. (cf KLKOHS). So $\Omega_{aa} = \alpha_{11}\mathcal{I}/\hbar$, $\Omega_{ab} = \alpha_{12}\mathcal{I}/\hbar$, $\Omega_{bb} = \alpha_{22}\mathcal{I}/\hbar$ (temporarily ignoring their detuning δ); and looking ahead to the definition of $f = \bar{\alpha}_{12}/2\hbar = \left(\alpha_{12} + \alpha_{21}\right)/2\hbar$ gives us $\Omega_{ab} = f\mathcal{I}$, noting $\mathcal{I} = 2A_qA_{q+1}^*$, compared to $\mathcal{I} = \frac{1}{2}E_qE_{q+1}^*$. Note that KLKOHS have defined $\Omega_{ab} = \frac{1}{2}\sum_q d_qE_qE_{q+1}^*$, so that $f = d_q$.

E. Two photon Bloch equations (cont)

An important difference between KLKOHS & HPB, and my equations is that my α_{ij} coupling parameters retain a "slow" time dependence at the Raman frequency W_{12}. I now turn my equations (2.3.5) into Bloch equations by first

working out d/dt of $c_1^*c_1$, $c_2^*c_2$, and $c_1^*c_2$,

$$\imath\hbar\frac{dc_1^*c_1}{dt} = \imath\hbar c_1^*\frac{dc_1}{dt} + \imath\hbar c_1\frac{dc_1^*}{dt} = -\alpha_{11}\mathfrak{I}c_1^*c_1 - \alpha_{12}\mathfrak{I}c_1^*c_2 + \alpha_{11}^*\mathfrak{I}c_1c_1^* + \alpha_{12}^*\mathfrak{I}c_1c_2^* \tag{2.5.1}$$

$$= +\alpha_{12}^*\mathfrak{I}c_1c_2^* - \alpha_{12}\mathfrak{I}c_1^*c_2 - [\alpha_{11} - \alpha_{11}^*]\,\mathfrak{I}c_1^*c_1 \tag{2.5.2}$$

$$= +\mathfrak{I}\left(\alpha_{12}^*c_1c_2^* - \alpha_{12}c_1^*c_2\right); \qquad \text{since} \quad \alpha_{11} = \alpha_{11}^* \tag{2.5.3}$$

$$\imath\hbar\frac{dc_2^*c_2}{dt} = \imath\hbar c_2^*\frac{dc_2}{dt} + \imath\hbar c_2\frac{dc_2^*}{dt} = -\alpha_{21}\mathfrak{I}c_2^*c_1 - \alpha_{22}\mathfrak{I}c_2^*c_2 + \alpha_{21}^*\mathfrak{I}c_2c_1^* + \alpha_{22}^*\mathfrak{I}c_2c_2^* \tag{2.5.4}$$

$$= -\alpha_{21}\mathfrak{I}c_2^*c_1 + \alpha_{21}^*\mathfrak{I}c_2c_1^* - [\alpha_{22} - \alpha_{22}^*]\,\mathfrak{I}c_2^*c_2 \tag{2.5.5}$$

$$= -\mathfrak{I}\left(\alpha_{21}c_2^*c_1 - \alpha_{21}^*c_2c_1^*\right); \qquad \text{since} \quad \alpha_{22} = \alpha_{22}^* \tag{2.5.6}$$

$$\imath\hbar\frac{dc_1c_2^*}{dt} = \imath\hbar c_1\frac{dc_2^*}{dt} + \imath\hbar c_2^*\frac{dc_1}{dt} = +\alpha_{21}^*\mathfrak{I}c_1c_1^* + \alpha_{22}^*\mathfrak{I}c_1c_2^* - \alpha_{11}\mathfrak{I}c_2^*c_1 - \alpha_{12}\mathfrak{I}c_2^*c_2 \tag{2.5.7}$$

$$= -\left(\alpha_{11} - \alpha_{22}^*\right)\mathfrak{I}c_2^*c_1 + \alpha_{21}^*\mathfrak{I}c_1c_1^* - \alpha_{12}\mathfrak{I}c_2^*c_2 \tag{2.5.8}$$

$$= -\left(\alpha_{11} - \alpha_{22}^*\right)\mathfrak{I}c_1c_2^* - \alpha_{12}\mathfrak{I}\left[c_2c_2^* - c_1^*c_1\right]; \qquad \text{since} \quad \alpha_{21}^* \approx \alpha_{12} \tag{2.5.9}$$

Use $\rho_{12} = c_1c_2^*$ and $w = c_2c_2^* - c_1^*c_1$,

$$\imath\hbar\frac{dw}{dt} = -\mathfrak{I}\left(\alpha_{21}c_2^*c_1 - \alpha_{21}^*c_2c_1^*\right) - \mathfrak{I}\left(\alpha_{12}^*c_1c_2^* - \alpha_{12}c_1^*c_2\right) \tag{2.5.10}$$

$$= \mathfrak{I}\left(-\alpha_{12}^*c_1c_2^* + \alpha_{21}^*c_1^*c_2 - \alpha_{21}c_1c_2^* + \alpha_{12}c_1^*c_2\right) \tag{2.5.11}$$

$$= -\mathfrak{I}\left(\alpha_{12}^* + \alpha_{21}\right)c_1c_2^* + \mathfrak{I}\left(\alpha_{21}^* + \alpha_{12}\right)c_1^*c_2 \tag{2.5.12}$$

$$= -\mathfrak{I}\left(\alpha_{12}^* + \alpha_{21}\right)\rho_{12} + \mathfrak{I}\left(\alpha_{21}^* + \alpha_{12}\right)\rho_{12}^* \tag{2.5.13}$$

$$\imath\hbar\frac{d\rho_{12}}{dt} = -\left(\alpha_{11} - \alpha_{22}^*\right)\mathfrak{I}c_1c_2^* - \alpha_{12}\mathfrak{I}\left[c_2c_2^* - c_1^*c_1\right] \tag{2.5.14}$$

$$= -\left(\alpha_{11} - \alpha_{22}^*\right)\mathfrak{I}\rho_{12} - \alpha_{12}\mathfrak{I}w \tag{2.5.15}$$

Hence

$$\frac{d\rho_{12}}{dt} = \imath\frac{\left(\alpha_{11} - \alpha_{22}^*\right)}{\hbar}\mathfrak{I}\rho_{12} + \imath\frac{\alpha_{12}}{\hbar}\mathfrak{I}w \tag{2.5.16}$$

$$\frac{dw}{dt} = +\imath\frac{\left(\alpha_{12}^* + \alpha_{21}\right)}{\hbar}\mathfrak{I}\rho_{12} - \imath\frac{\left(\alpha_{21}^* + \alpha_{12}\right)}{\hbar}\mathfrak{I}\rho_{12}^*, \tag{2.5.17}$$

$$= +\imath\frac{2\alpha_{12}^*}{\hbar}\mathfrak{I}\rho_{12} - \imath\frac{2\alpha_{12}}{\hbar}\mathfrak{I}\rho_{12}^*. \tag{2.5.18}$$

F. Transformations of the Bloch Equations

I define a new coupling parameter f' (c.f. ω_B), following the definition of $\bar{\alpha}_{12}$ in eqn.(2.2.6), and $\omega_b = W_2 - W_1 = W_{21}$. I also define a ω_A, which corresponds to the intensity dependent shift detuning shift proportional to $(\alpha_{22} - \alpha_{11})$. Thus, factoring any complex phase of $\bar{\alpha}_{12}$ into the angle δ', I get –

$$f'e^{-\imath\omega_b t} = \frac{\alpha_{12}}{\hbar} \simeq \frac{\bar{\alpha}_{12}}{2\hbar}e^{-\imath\omega_b t - \imath\delta'}; \qquad \alpha_{12} = \frac{1}{\hbar}e^{-\imath\omega_b t}\sum_j d_{ij}d_{j2}\frac{-W_{j2}}{W_{j2}^2 - \omega_0^2} \tag{2.6.1}$$

$$g' = \frac{\alpha_{11} - \alpha_{22}^*}{\hbar} \qquad \text{NB: this is a real quantity, see eqns. (2.2.11, 2.2.17)} \tag{2.6.2}$$

$$\omega_B = \frac{\left(\alpha_{12} + \alpha_{21}^*\right)}{2\hbar}\mathfrak{I} \quad = \frac{\bar{\alpha}_{12}}{2\hbar}e^{-\imath\omega_b t - \imath\delta'}\mathfrak{I} \quad = f'e^{-\imath\omega_b t - \imath\delta'}\mathfrak{I} \tag{2.6.3}$$

$$\omega_A = \frac{\left(\alpha_{11} - \alpha_{22}^*\right)}{\hbar}\mathfrak{I}. \tag{2.6.4}$$

The Bloch equations (2.5.16, 2.5.17) can now be rewritten –

$$\frac{d\rho_{12}}{dt} = \imath g'\mathfrak{I}\rho_{12} + \imath f'\mathfrak{I}we^{-\imath\omega_b t - \imath\delta'} \tag{2.6.5}$$

$$\frac{dw}{dt} = +2\imath f'\mathfrak{I}\left[\rho_{12}e^{+\imath\omega_b t + \imath\delta'} - \rho_{12}^*e^{-\imath\omega_b t - \imath\delta'}\right]. \tag{2.6.6}$$

In analogy to both the standard two level atom Bloch equations, and those in HPB (HPB 18), these equations can have losses introduced, in –

$$\frac{d\rho_{12}}{dt} = -\gamma_2\rho_{12} + \imath g'\mathcal{I}\rho_{12} + \imath f'\mathcal{I}we^{-\imath\omega_b t-\imath\delta'} \tag{2.6.7}$$

$$\frac{dw}{dt} = -\gamma_1\left(w-w_i\right) + 2\imath f'\mathcal{I}\left[\rho_{12}e^{+\imath\omega_b t+\imath\delta'} - \rho_{12}^* e^{-\imath\omega_b t-\imath\delta'}\right]. \tag{2.6.8}$$

In the standard atom-field case, the atom and field frequencies are similar, so an atom carrier could be chosen to match its evolution the field carrier frequency, leading naturally to a detuning term. However, in the Raman situation, the atomic frequency is far removed from the field frequency, so the frequency evolution that appears in the definition of ψ (eqn.(2.1.3)) is sufficient.

Here I define two things (a) new u, v variables that represent the density matrix element ρ_{12}, and (b) allow for a "detuning" rotation in ρ_{12}. This detuning rotation looks rather like a carrier+envelope representation for ρ_{12}, but without a "+c.c." since ρ_{12} is a complex quantity. There is no need for any kind of carrier+envelope transformation for the inversion w.

So,

$$\rho_{12} = \frac{u}{2} + \imath\frac{v}{2} = \rho'_{12}\exp\left(-\imath\Delta t - \imath\delta'\right) = \frac{u'}{2} + \imath\frac{v'}{2} \tag{2.6.9}$$

I now adapt the Bloch equations to allow for the rotation in ρ_{12}, and introduce a detuned transition frequency $\omega'_b = \omega_b - \Delta$,

$$\partial_t\left(\rho'_{12}e^{-\imath\Delta t-\imath\delta'}\right) = -\imath\Delta\rho_{12}e^{-\imath\Delta t-\imath\delta'} + e^{-\imath\Delta t-\imath\delta'}\partial_t\rho'_{12} \tag{2.6.10}$$

$$= -\gamma_2\rho'_{12}e^{-\imath\Delta t-\imath\delta'} + \imath g'\mathcal{I}\rho'_{12}e^{-\imath\Delta t-\imath\delta'} + \imath f'\mathcal{I}we^{-\imath\omega_b t-\imath\delta'} \tag{2.6.11}$$

$$\partial_t w = -\gamma_1\left(w-w_i\right) + 2\imath f'\mathcal{I}\left(\rho'_{12}e^{-\imath\Delta t-\imath\delta'}e^{+\imath\omega_b t+\imath\delta'} - \rho'^*_{12}e^{+\imath\Delta t+\imath\delta'}e^{-\imath\omega_b t-\imath\delta'}\right) \tag{2.6.12}$$

$$\Longrightarrow \quad e^{-\imath\Delta t-\imath\delta'}\partial_t\rho'_{12} = \left(-\gamma_2+\imath\Delta\right)\rho'_{12}e^{-\imath\Delta t-\imath\delta'} + \imath g'\mathcal{I}\rho'_{12}e^{-\imath\Delta t-\imath\delta'} + \imath f'\mathcal{I}we^{-\imath\omega_b t-\imath\delta'} \tag{2.6.13}$$

$$\partial_t w = -\gamma_1\left(w-w_i\right) + 2\imath f'\mathcal{I}\left(\rho'_{12}e^{+\imath\omega'_b t} - \rho'^*_{12}e^{-\imath\omega'_b t}\right) \tag{2.6.14}$$

$$\Longrightarrow \quad \partial_t\rho'_{12} = \left(-\gamma_2+\imath\Delta\right)\rho'_{12} + \imath g'\mathcal{I}\rho'_{12} + \imath f'\mathcal{I}e^{\imath\omega'_b t} \tag{2.6.15}$$

$$\partial_t w = -\gamma_1\left(w-w_i\right) + 2\imath f'\mathcal{I}\left(\rho'_{12}e^{+\imath\omega'_b t} - \rho'^*_{12}e^{-\imath\omega'_b t}\right) \tag{2.6.16}$$

Notice that we have made the fixed complex phase (the δ') vanish from the equations; this is not dependent on the presence of a finite "detuning" Δ. In what follows, $\Delta = 0$, and there are two cases depending on the chosen meaning for $\mathcal{I}$; however note that *both give the same result.*

CASE (i): $\mathcal{I} = E^2$: continue by applying the carrier+envelope for the E field $E = Ae^{-\imath\omega_0 t} + A^*e^{+\imath\omega_0 t}$.

CASE (ii): $\mathcal{I} = 2A^*A$: we can jump straight to eqn.(2.6.25, 2.6.26) (i.e. (D)), since the RWA was made when deriving the couplings α_{nj}.

The CASE (i) $\mathcal{I} = E^2$ starting point is –

$$\partial_t \rho'_{12} = (-\gamma_2 + \imath\Delta + \imath g'\mathcal{I})\,\rho'_{12} \quad + \imath f'\mathcal{I} w e^{-\imath\omega'_b t} \tag{2.6.17}$$

$$\partial_t w = -\gamma_1 (w - w_i) \quad + 2\imath f'\mathcal{I}\left(\rho'_{12} e^{+\imath\omega'_b t} - \rho'^{*}_{12} e^{-\imath\omega'_b t}\right), \tag{2.6.18}$$

$$(B) \Longrightarrow \qquad \partial_t \rho'_{12} = \left(-\gamma_2 + \imath\Delta + \imath g' E^2\right)\rho'_{12} \quad \imath f' E^2 e^{-\imath\omega'_b t}.w \tag{2.6.19}$$

$$\partial_t w = -\gamma_1 (w - w_i) \quad + 2\imath f' E^2\left(\rho'_{12} e^{+\imath\omega'_b t} - \rho'^{*}_{12} e^{-\imath\omega'_b t}\right), \tag{2.6.20}$$

$$(C) \Longrightarrow \qquad \partial_t \rho'_{12} = \left(-\gamma_2 + \imath\Delta + \imath g' E^2\right)\rho'_{12} \quad + \imath f'\left[A e^{-\imath\omega_0 t} + A^* e^{+\imath\omega_0 t}\right]^2 w e^{-\imath\omega'_b t} \tag{2.6.21}$$

$$\partial_t w = -\gamma_1 (w - w_i) \quad + 2\imath f'\left[A e^{-\imath\omega_0 t} + A^* e^{+\imath\omega_0 t}\right]^2\left(\rho_{12} e^{+\imath\omega'_b t} - \rho^*_{12} e^{-\imath\omega'_b t}\right) \tag{2.6.22}$$

(apply a RWA about ω_0) $\qquad :::\ \left[A e^{-\imath\omega_0 t} + A^* e^{+\imath\omega_0 t}\right]^2$

$$= \left[A^2 e^{-2\imath\omega_0 t} + 2A^* A + A^{*2} e^{+2\imath\omega_0 t}\right] \tag{2.6.23}$$

$$\Longrightarrow \qquad \approx 2A^* A \tag{2.6.24}$$

$$CASE(ii)\ \&\ (D) \Longrightarrow \qquad \partial_t \rho'_{12} = (-\gamma_2 + \imath\Delta + 2\imath g' A^* A)\,\rho'_{12} \quad + 2\imath f' A^* A w e^{-\imath\omega'_b t} \tag{2.6.25}$$

$$\partial_t w = -\gamma_1 (w - w_i) \quad + 4\imath f' A^* A\left(\rho_{12} e^{+\imath\omega'_b t} - \rho^*_{12} e^{-\imath\omega'_b t}\right), \tag{2.6.26}$$

$$(E) \qquad \partial_t \rho'_{12} = (-\gamma_2 + \imath\Delta + 2\imath g' A^* A)\,\rho'_{12} + 2\imath f' A^* A w.e^{-\imath\omega'_b t} \tag{2.6.27}$$

$$\partial_t w = -\gamma_1 (w - w_i) + 4\imath f' A^* A\left(\rho'_{12} e^{+\imath\omega'_b t} - \rho'^{*}_{12} e^{-\imath\omega'_b t}\right), \tag{2.6.28}$$

$$(F : \text{split}\ \ \rho_{12},) \Longrightarrow \qquad \partial_t u' = -\gamma_2 u' - (\Delta + 2g' A^* A)\,v' \quad + 4f' A^* A w.\sin(\omega'_b t) \tag{2.6.29}$$

$$\partial_t v' = -\gamma_2 v' + (\Delta + 2g' A^* A)\,u' \quad + 4f' A^* A w.\cos(\omega'_b t) \tag{2.6.30}$$

$$\partial_t w = -\gamma_1 (w' - w_i) \quad - 4f' A^* A u'.\sin(\omega'_b t) \quad - 4f' A^* A v'.\cos(\omega'_b t), \tag{2.6.31}$$

Note $2\rho_{12} = u + \imath v$, since $\rho_{12} = c_1 c_2^*$, as per eqn.(2.6.9); this means the 2 in the u, v equations becomes 4, whereas the 4 in the w equation isn't doubled to 8. Note (again) that the Stark shift parameter g' is real valued.

1. *Rotations*

Looking at eqns.(2.6.29, 2.6.30, 2.6.31), we can see three rotation angles: $\theta_{uv} = \Delta$, $\theta_{uw} = 4f' A^* A \sin(\omega_b t - \delta')$, and $\theta_{vw} = 4f' A^* A \cos(\omega_b t - \delta')$, which apply to the coordinate pairs (u', v'), (u', w), and (v', w) respectively. For extra generality, I will allow a complex valued $f' = f_r + \imath f_i$, but note that previous definitions make f' real.)

In vector notation, ignoring the losses, we can construct a torque vector $\vec{\Omega}$ (unchecked signs),

$$\partial_t u' = 0.u' - \theta_{uv}.v' + \theta_{uw}.w \tag{2.6.32}$$

$$\partial_t v' = \Delta.u' + 0.v' - \theta_{vw}.w \tag{2.6.33}$$

$$\partial_t w = -\theta_{uw}.u' + \theta_{vw}.v' + 0.w \tag{2.6.34}$$

$$\frac{d}{dt}[u', v', w] = [-4f'_r A^* A, \quad -4f'_i A^* A, \quad \Delta] \tag{2.6.35}$$

$$= [\theta_{vw},\ \theta_{uw},\ \theta_{uv}] \times [u', v', w] \tag{2.6.36}$$

$$\Longrightarrow \qquad \frac{d}{dt}\vec{U} = \vec{\Omega} \times \vec{U} \tag{2.6.37}$$

I should now be able to turn this into a (unitary) rotation matrix for the $\vec{U}$ vector.

G. The polarization driving the field

There is a distinction between the atomic polarization P, and the effect on the field of that atomic polarization. This is beause we are dealing with a nonlinear interaction. I denote the (effective) Raman polarization $\mathcal{P}$, and this

quantity is the one that appears in the wave equation. Allen and Eberly *"Optical resonance and two level atoms"* [15] have, for the standard (non-Raman) case (skipping the integral), an eqn.(AE 4.2) –

$$(AE\ 4.2) \qquad P(t,z) \ = \ \mathcal{N}d\left[u\cos(\omega t - Kz) - v\sin(\omega t - Kz)\right] \tag{2.7.1}$$

HPB, after summing the electric field components (which absorbs a 1/2), have

$$(HPB\ 21) \qquad P \ = \ \frac{1}{4}e^{-\imath\theta}\left(u+\imath v\right)\sigma\alpha_{12}\sum_j\left[V_j e^{\imath\omega_{j-1}t} + V_j^* e^{-\imath\omega_{j+1}t}\right] \quad + c.c. \tag{2.7.2}$$

$$= \ \frac{1}{2}e^{-\imath\theta}\left(u+\imath v\right)\sigma\alpha_{12}e^{-\imath\omega_b t}\frac{1}{2}\sum_j\left[V_j e^{\imath\omega_j t} + V_j^* e^{-\imath\omega_j t}\right] \quad + c.c. \tag{2.7.3}$$

$$= \ \frac{1}{2}e^{-\imath\theta}\left(u+\imath v\right)\sigma\alpha_{12}e^{-\imath\omega_b t}E \quad + c.c. \tag{2.7.4}$$

I now write down the the polarization envelope B, in my variables, and based on the same carrier as the electric field. I also introduce a complex factor equivalent to HPB's $e^{-\imath\theta}$, for closer matching of the expressions.

$$\mathcal{P} \ = \mathcal{B}e^{-\imath\Xi} + \mathcal{B}^* e^{+\imath\Xi} \ = \ \rho_{12}e^{-\imath\theta}\sigma\alpha_{12}\left(Ae^{-\imath\Xi} + A^* e^{+\imath\Xi}\right) \quad + c.c. \tag{2.7.5}$$

$$= \ \zeta\rho'_{12}e^{-\imath\theta}\sigma.\bar{\alpha}_{12}e^{-\imath\omega'_b t}\left(Ae^{-\imath\Xi} + A^* e^{+\imath\Xi}\right) \quad + c.c. \tag{2.7.6}$$

$$= \ \zeta\rho'_{12}e^{-\imath\theta}\sigma.\bar{\alpha}'_{12}e^{-\imath\omega'_b t}\left(Ae^{-\imath\Xi} + A^* e^{+\imath\Xi}\right) + \zeta{\rho'_{12}}^* e^{+\imath\theta}\sigma.\bar{\alpha}'_{12}e^{+\imath\omega'_b t}\left(A^* e^{+\imath\Xi} + Ae^{-\imath\Xi}\right) \tag{2.7.7}$$

$$= \ \zeta\rho'_{12}e^{-\imath\theta}\sigma.\bar{\alpha}'_{12}e^{+\imath\omega'_b t}Ae^{-\imath\Xi} \quad + \zeta{\rho'_{12}}^* e^{+\imath\theta}\sigma.\bar{\alpha}'_{12}e^{-\imath\omega'_b t}Ae^{-\imath\Xi}$$
$$+\zeta\rho'_{12}e^{-\imath\theta}\sigma.\bar{\alpha}'_{12}e^{+\imath\omega'_b t}A^* e^{+\imath\Xi} \quad + \zeta{\rho'_{12}}^* e^{+\imath\theta}\sigma.\bar{\alpha}'_{12}e^{-\imath\omega'_b t}A^* e^{+\imath\Xi} \tag{2.7.8}$$

$$= \ \zeta\sigma.\bar{\alpha}'_{12}\left\{\rho'_{12}e^{+\imath\omega'_b t-\imath\theta} + {\rho'_{12}}^* e^{-\imath\omega'_b t+\imath\theta}\right\}Ae^{-\imath\Xi}$$
$$+\zeta\sigma.\bar{\alpha}'_{12}\left\{\rho'_{12}e^{+\imath\omega'_b t-\imath\theta} + {\rho'_{12}}^* e^{-\imath\omega'_b t+\imath\theta}\right\}A^* e^{+\imath\Xi} \tag{2.7.9}$$

$$= \ \zeta\sigma.\bar{\alpha}'_{12}X(t)Ae^{-\imath\Xi} + \zeta\sigma.\bar{\alpha}'_{12}X(t)A^* e^{+\imath\Xi} \tag{2.7.10}$$

where the real valued is $X(t) \ = \ \left\{\rho'_{12}e^{+\imath\omega'_b t-\imath\theta} + {\rho'_{12}}^* e^{-\imath\omega'_b t+\imath\theta}\right\}$ (2.7.11)

$$= \ \left\{\frac{1}{2}\left(u'+\imath v'\right)e^{+\imath\omega'_b t-\imath\theta} + \frac{1}{2}\left(u'-\imath v'\right)e^{-\imath\omega'_b t+\imath\theta}\right\} \tag{2.7.12}$$

$$= \ \left\{u'\cos\left(\omega'_b t-\theta\right) - v'\sin\left(\omega'_b t-\theta\right)\right\} \tag{2.7.13}$$

and $\mathcal{B}(t) \ = \ \zeta\sigma\bar{\alpha}'_{12}X(t)A(t)$ (2.7.14)

From the post-ω_0 RWA at eqns.(2.6.29, 2.6.30, 2.6.31); above; and an un-scaled eqn. (FCPP 3.48), –

$$\partial_t u' \ = \ -\gamma_2 u' - \left(\Delta + 2g'A^*A\right)v' \quad + 4f'A^*Aw\sin(\omega'_b t) \tag{2.7.15}$$

$$\partial_t v' \ = \ -\gamma_2 v + \left(\Delta + 2g'A^*A\right)u' \quad + 4f'A^*Aw\cos(\omega'_b t) \tag{2.7.16}$$

$$\partial_t w \ = \ -\gamma_1\left(w' - w_i\right) \quad - 4f'A^*A.\sin(\omega'_b t)u' \quad - 4f'A^*A.\cos(\omega'_b t)v', \tag{2.7.17}$$

$$\partial_z A(t) \ = \ \frac{2\imath\pi\omega_0}{c_0 n_0}\frac{\mathcal{B}(t)}{4\pi\epsilon_0}, \qquad \text{NB: units } [A]/m = \frac{s^{-1}}{m.s^{-1}}[\mathcal{B}] \ \rightarrow [A] = [\chi][B][A] \ \rightarrow [\chi] = [B^{-1}] = 1 \tag{2.7.18}$$

$$= \ \frac{\imath\omega_0}{2c_0 n_0\epsilon_0}\mathcal{B}(t) \tag{2.7.19}$$

$$= \ \imath\frac{\zeta\sigma\bar{\alpha}_{12}\omega_0}{2c_0 n_0\epsilon_0}\ .\ \left[u'\cos\left(\omega'_b t-\theta\right) - v'\sin\left(\omega'_b t-\theta\right)\right]\ .\ A(t) \tag{2.7.20}$$

$$= \ \imath\frac{\zeta\sigma\bar{\alpha}_{12}\omega_0}{2c_0 n_0\epsilon_0}\ .\ A(t)X(t) \tag{2.7.21}$$

Comparing the prefactor of eqn.(2.7.21) to that of the corrected (HPB 22),

$$(\text{HPB } 22) \qquad \frac{\partial V_j}{\partial z} \ = \ \frac{\sigma\alpha_{12}}{4\epsilon_0 c}\omega_j\left[e^{-\imath\theta}\left(u-\imath v\right)V_{j+1} - e^{+\imath\theta}\left(u+\imath v\right)V_{j-1}\right] \tag{2.7.22}$$

we can see that the only apparent differences are a factor of n_0, and in that HPB have a $e^{\pm\imath\theta}$ term that I omit. By imagining the cosine term split up into + and − frequency exponentials, it is easy to see how the relations between V_j and $V_{j\pm1}$ will arise. Note the appearance (in HPB) of a carrier-dependent ω_i term, which in a standard multi-field approach would lead to different prefactors on the different $\partial_\xi A_i$ equations.

1. Simulation "photon" variables

Scale the field E into square-root intensity "photon" variables, from $I = 2n_0c_0\epsilon_0E^2$, so that

$$A_p(t) \;=\; \sqrt{2c_0n_0\epsilon_0}\;.\;A(t) \tag{2.7.23}$$

$$f_p \;=\; \frac{f'}{2c_0n_0\epsilon_0}; \qquad g_p \;=\; \frac{g'}{2c_0n_0\epsilon_0}; \tag{2.7.24}$$

Hence $f'A^*A \longrightarrow f_pA_p^*A_p$, so that

$$\partial_t u' \;=\; -\gamma_2 u' - (\Delta + 2g_pA^*A)\,v' \quad + 4f_pA_p^*A_p w\sin(\omega_b' t) \tag{2.7.25}$$

$$\partial_t v' \;=\; -\gamma_2 v + (\Delta + 2g_pA^*A)\,u' \quad + 4f_pA_p^*A_p w\cos(\omega_b' t) \tag{2.7.26}$$

$$\partial_t w \;=\; -\gamma_1\,(w' - w_i) \quad - 4f_pA_p^*A_p u'\sin(\omega_b' t) \quad - 4f_pA_p^*A_p v'\cos(\omega_b' t) \tag{2.7.27}$$

$$\partial_z A_p(t) \;=\; \imath\frac{\zeta\sigma\bar{\alpha}_{12}\omega_0}{2c_0n_0\epsilon_0}A_p(t)X(t) \tag{2.7.28}$$

$$\;=\; \imath\zeta\sigma\omega_0\frac{4\hbar c_n n_0\epsilon_0 f_p}{2c_0n_0\epsilon_0}A_p(t)X(t) \tag{2.7.29}$$

$$\;=\; \imath\frac{\zeta}{2}\,(4\sigma\hbar)\,\omega_0 f_pA_p(t)X(t) \qquad = \imath\frac{\zeta}{2}\omega_0 R f_pA_p(t)X(t); \quad R = 4\sigma\hbar \tag{2.7.30}$$

since

$$\bar{\alpha}_{12} = 2\hbar f' \quad \rightarrow \frac{\bar{\alpha}_{12}}{2c_0n_0\epsilon_0} = 2\hbar f_p \quad \rightarrow \bar{\alpha}_{12} = 4\hbar c_0n_0\epsilon_0 f_p \tag{2.7.31}$$

Scalings:
$\hbar = 1.05\times10^{-34}\text{Js} \rightarrow 1.05\times10^{-10}\text{nJ.fs}$
$\sigma = X\text{m}^{-3} \rightarrow X\times10^{-18}\mu\text{m}^{-3}$
$2c_0n_0\epsilon_0 = 5.31\times10^{-3}\text{m.s}^{-1}\;.\;\text{J.m}^{-1}.\text{V}^{-2} \rightarrow 5.31\times10^{-3}\;.\;\text{J.s}^{-1}.\text{V}^{-2} \rightarrow 5.31\times10^{-9}\;.\;\text{nJ.fs}^{-1}.\text{V}^{-2}$
Scalings:
$[c\epsilon_0] \;=\; m.s^{-1}\;.\;J.m^{-1}.V^{-2} \;=\; J.s^{-1}.C^2.J^{-2} \;=\; C^2.J^{-1}.s^{-1} \;=\; C^2\;.\;10^{-9}nJ^{-1}\;.\;10^{-15}fs^{-1} \;=\; 10^{-24}.C^2.nJ^{-1}.fs^{-1}$
$[\epsilon_0] = J.m^{-1}.V^{-2} \;= J.m^{-1}.C^2.J^{-2} \;= C^2.J^{-1}.m^{-1} = C^2\;.\;10^{-9}nJ^{-1}\;.\;10^{-6}\mu m^{-1} \;= 10^{-15}.C^2.nJ^{-1}.\mu m^{-1}$

2. Comments on the field propagation

We can see that the derivative of A is proportional to $\imath A$, hence the (time-domain) evolution just rotates each point $A(t)$ differently without changing its amplitude. This might seem to imply that we will never get a shorter pulse than we put in; but note that the A is an *envelope*, and the field is $A + A^*$, so that in principle the phase of A might be such that its amplitudes cancel in certain t regions but not others, leading to a shorter pulse (and the magnetic field H also, since the fields are plane polarized). However, it seems unlikely that this will happen (barring some miraculous coincidence) from purely Raman effects.

If I can predict the output spectral phases though, a structure with a suitably designed dispersion *might* be able to impose the desired phases. It would only be necessary to get the dispersion right at the comb points of the spectrum. See Shverdin et.al. [16], who do a four-wave mixing optimization procedure in their experiment to match their phases appropriately.

H. The steady state and gain co-efficient

Starting from the eqns.(2.7.15, 2.7.16, 2.7.17), assume v' is steady state (NB $\gamma_2 = 1/T_2$), so (with $w = -1$) –

$$\partial_t \approx 0 \qquad = -\gamma_2 v' + 4f'A^*A(-1)\cos(\omega_b t) \tag{2.8.1}$$

$$\Longrightarrow \qquad v_0' \;=\; \frac{4f'}{\gamma_2}\;.\;A^*A.\cos(\omega_b t) \qquad = 4f'T_2\;.\;A^*A.\cos(\omega_b t) \tag{2.8.2}$$

I now use the field propagation equation (2.7.30), with the A field envelope on each side scaled into "photon" variables. Inserting the steady state of v' calculated immediately above, we have –

$$\begin{aligned}
\partial_z A_p &= \imath \frac{\zeta \sigma \bar{\alpha}_{12} \omega_0}{2 c_0 n_0 \epsilon_0} A_p v'_0 \sin(\omega_b t) && (2.8.3)\\
&= \imath \frac{\zeta \sigma \bar{\alpha}_{12} \omega_0}{2 c_0 n_0 \epsilon_0} A_p \,.\, 4 f' T_2 \,.\, A^* A . \cos(\omega_b t) . \sin(\omega_b t) && (2.8.4)\\
&= \imath \frac{\zeta \sigma \bar{\alpha}_{12} \omega_0}{2 c_0 n_0 \epsilon_0} A_p \,.\, 4 T_2 \frac{\bar{\alpha}_{12}}{2\hbar} \frac{A_p^* A_p}{2 c_0 n_0 \epsilon_0} [\cos(\omega_b t) . \sin(\omega_b t)] && (2.8.5)\\
&= \imath \frac{\zeta \sigma \omega_0 T_2 \bar{\alpha}_{12}^2}{2 c_0^2 n_0^2 \epsilon_0^2 \hbar} A_p A_p^* A_p . [\cos(\omega_b t) . \sin(\omega_b t)] && (2.8.6)\\
&= \imath G_p A_p A_p^* A_p . [\cos(\omega_b t) . \sin(\omega_b t)] , && (2.8.7)\\
G_p &= \frac{\zeta}{2} \frac{\sigma \omega_0 T_2 \bar{\alpha}_{12}^2}{c_0^2 n_0^2 \epsilon_0^2 \hbar} . && (2.8.8)
\end{aligned}$$

Apparently, therefore, this G_p corresponds to the usual "gain co-efficient" g:

$$\text{cf (SMN)} \qquad g' = \frac{\sigma \omega_0 T_2 \alpha_{12}^2}{c^2 \epsilon_0^2 \hbar}, \tag{2.8.9}$$

and we can assume that they are identical but for a factor of $2n_0^2/\zeta$. I do not know whether SMN silently assumes $n_0 = 1$, or factors n_0 into c. In any case $n_0 = 1$ is usually accurate enough in gases.

However, one hidden complication with my above equation is that A_p includes both the center "pump" field and the Raman sideband we are amplifying – thus to properly check the gain co-efficient, we should expand it into its components, as is done below.

1. Pump and sideband calculation

A_p includes both fundamental A_1 and its Raman sideband $A_2 = A'_2 e^{-\imath \omega_b t}$. For the moment, I leave the calculation in a rather abbreviated state – probably it should be shifted to the Multi-field section following later. So

$$\begin{aligned}
A_p A_p^* A_p &= [A_1 + A_2] [A_1^* + A_2^*] [A_1 + A_2] && (2.8.10)\\
&= [A_1 A_1^* + A_1 A_2^* + A_2 A_1^* + A_2 A_2^*] [A_1 + A_2] && (2.8.11)\\
&= A_1 A_1^* A_1 + A_1 A_2^* A_1 + A_2 A_1^* A_1 + A_2 A_2^* A_1 + A_1 A_1^* A_2 + A_1 A_2^* A_2 + A_2 A_1^* A_2 + A_2 A_2^* A_2 && (2.8.12)\\
\text{1st order terms in } A_2 \text{ only:} \quad &\simeq A_1^2 A_2^* + 2 A_2 A_1^* A_1 && (2.8.13)\\
\text{drop counter rotating } A_1^2 A_2^* \text{:} \quad &\simeq 2 A_2 A_1^* A_1 . && (2.8.14)
\end{aligned}$$

Thus the sideband envelope A'_2 evolves as

$$\begin{aligned}
\partial_z A'_2 &= \imath G_p A'_2 . 2 A_1^* A_1 . \cos(\omega_b t) && (2.8.15)\\
&= \imath G_p A'_2 . I_1 . \cos(\omega_b t) && (2.8.16)\\
&= \imath G'_p A'_2 . I_1 . \cos(\omega_b t), && (2.8.17)\\
\text{where} \qquad G'_p &= \frac{\zeta}{2} \frac{\sigma \omega_0 T_2 \bar{\alpha}_{12}^2}{c_0^2 n_0^2 \epsilon_0^2 \hbar} = G_p && (2.8.18)
\end{aligned}$$

since $I_m = \frac{1}{2} A_m^* A_m$. I don't convert A'_2 into I_2 because it occurs equally on both side of the equation.

III. MULTI-FIELD VARIANT OF SINGLE-FIELD RAMAN THEORY

The single-field Raman model above can be converted into a traditional multi-field model as developed in e.g. HPB [11] or Syed, McDonald and New [17] by replacing the field envelope with a sum of multiple envelopes using carrier exponentials spaced at the Raman frequency. When doing this, I will only get the correct multi-field form if few-cycle

(either SEWA or GFEA) corrections to the field evolution part of the theory are applied to the effective polarization caused by the Raman transition.

The idea is to replace the single field envelope with a sum of multiple envelopes spaced at the Raman frequency, which are best placed to represent the comb of frequencies generated by the Raman interaction. Note that it will not necessarily be identical to HPB and/or SMN, because the field equations are derived from a propagation equation using a ω_0, β_0 carrier, but it will be very closely related.

Starting from eqns.(2.6.27,2.6.28), Since the single-field evolution equation (eqn.(2.7.21)) uses an envelope A that is based on a carrier (see eqn.(2.1.21)), the single-field envelope A is replaced with A_j's at frequency $\omega_j = \omega_0 + j\omega_b$ with wavevector $k_j = k(\omega_j)$; thus $\omega'_j = \omega_j - \omega_0$, $k'_j = k_j - k(\omega_0) = k_j - k_0$; also $\beta \leftrightarrow k$. The single-field envelope in terms of the new A_j's is

$$A = \sum_j A_j \exp\left[-\imath\left(\omega_j t - k_j z\right)\right] \tag{3.0.1}$$

A. Polarization (ρ'_{12})

First I will handle the polarization (ρ'_{12}) equation (from eqn.(2.6.25)) (watch for any $\omega'_b = \omega_b - \Delta$ confusion, and note $2\rho'_{12} = u' + \imath v'$) –

$$\partial_t \rho'_{12} = \left(-\gamma_2 + \imath\Delta + 2\imath g' A^* A\right)\rho'_{12} + 2\imath f' A^* A w e^{-\imath\omega'_b t} \tag{3.1.1}$$

$$(A1) \implies \partial_t \rho'_{12} = \left(-\gamma_2 + \imath\Delta\right)\rho'_{12} + 2\imath g'\rho'_{12}\sum_j\sum_k A^*_j A_k e^{+\imath(\omega_j-\omega_k)t} e^{+\imath(-k_j+k_k)z}$$
$$+ 2\imath f'\sum_j\sum_k A^*_j A_k w e^{+\imath\left(\omega_j-\omega_k-\omega'_b\right)t} e^{+\imath(-k_j+k_k)z} \tag{3.1.2}$$

$$= \left(-\gamma_2 + \imath\Delta\right)\rho'_{12} + 2\imath g'\rho'_{12}\sum_j\sum_k A^*_j A_k e^{+\imath(j-k)\omega_b t} e^{+\imath(-k_j+k_k)z}$$
$$+ 2\imath f'\sum_j\sum_k A^*_j A_k w e^{+\imath(j-1-k)\omega_b t+\imath\Delta t} e^{+\imath(k_k-k_j)z} \tag{3.1.3}$$

$$(RWA) \approx \left(-\gamma_2 + \imath\Delta\right)\rho'_{12} + 2\imath g'\rho'_{12}\sum_j A^*_j A_j w + 4\imath f'\sum_j A^*_j A_{j-1}.w.e^{+\imath\Delta t}.e^{+\imath(k_{j+1}-k_j)z} \tag{3.1.4}$$

$$\approx \left(-\gamma_2 + \imath\Delta + 2\imath g'\sum_j A^*_j A_j\right)\rho'_{12} + 4\imath f'\sum_j A_j A^*_{j+1}.w.e^{+\imath\Delta t}.e^{+\imath(k_j-k_{j-1})z} \tag{3.1.5}$$

$$\approx \left(-\gamma_2 + \imath\Delta + 2\imath g'\sum_j A^*_j A_j\right)\rho'_{12} + 4\imath f' w.\mathbb{R}e\left[\sum_j A_j A^*_{j+1}.e^{+\imath\Delta t}.e^{+\imath(k_j-k_{j-1})z}\right]$$
$$+ 4\imath^2 f' w.\mathbb{I}m\left[\sum_j A_j A^*_{j+1}.e^{+\imath\Delta t}.e^{+\imath(k_j-k_{j-1})z}\right] \tag{3.1.6}$$

$$(\text{split } \rho'_{12}) \quad \partial_t u = -\gamma_2 u - \left(\Delta + 2g'\sum_j A^*_j A_j\right) v - 8f' w.\mathbb{I}m\left[\sum_j A_j A^*_{j+1}.e^{+\imath\Delta t}.e^{+\imath(k_j-k_{j-1})z}\right] \tag{3.1.7}$$

$$\partial_t v = -\gamma_2 v + \left(\Delta + 2g'\sum_j A^*_j A_j\right) u + 8f' w.\mathbb{R}e\left[\sum_j A_j A^*_{j+1}.e^{+\imath\Delta t}.e^{+\imath(k_j-k_{j-1})z}\right] \tag{3.1.8}$$

Where the factor $2\imath f'$ turns into $4\imath f'$ because the double summation gives two identical terms that only occur once in the single summation. When split into equations for u' and v', the corresponding factor becomes $8\imath f'$. This equation for ρ'_{12} is equivalent to (SMN 2)[17], except I have just $k_j - k_{j-1}$ whereas they have $\Delta_j = k_j - k_{j-1} - k_0 + k_{-1}$; however note they have the reverse sign in their carrier wave, so the only physical difference is the $k_0 - k_{-1}$ part; also my definitions of the coupling differs slightly.

B. Inversion (w)

And now the inversion (w) equation (also from eqn.(2.6.25)) (watch for any ω_b vs ω'_b confusion) –

$$\partial_t w = -\gamma_1 (w - w_i) + 4\imath f' A^* A \left[\rho'_{12} e^{+\imath\omega'_b t} - \rho'^*_{12} e^{-\imath\omega'_b t}\right] \tag{3.2.1}$$

$$(A2)\quad \partial_t w = -\gamma_1 (w - w_i) + 4\imath f' \sum_j \sum_k A^*_j A_k . e^{+\imath(\omega_j - \omega_k)t} . e^{+\imath(-k_j + k_k)z} \left[\rho'_{12} e^{+\imath\omega'_b t} - \rho'^*_{12} e^{-\imath\omega'_b t}\right] \tag{3.2.2}$$

$$= -\gamma_1 (w - w_i) + 4\imath f' \sum_j \sum_k \left[A^*_j A_k \rho'_{12} e^{+\imath(j-k+1)\omega_b t - \imath\Delta t} - A^*_j A_k \rho'^*_{12} e^{+\imath(j-k-1)\omega_b t + \imath\Delta t}\right] . e^{+\imath(k_k - k_j)z} \tag{3.2.3}$$

$$(RWA)\quad \approx -\gamma_1 (w - w_i) + 8\imath f' \sum_j \left[A^*_j A_{j+1} \rho'_{12} . e^{-\imath\Delta t} . e^{+\imath(k_{j+1} - k_j)z} - A^*_j A_{j-1} \rho'^*_{12} . e^{+\imath\Delta t} . e^{+\imath(k_{j-1} - k_j)z}\right] \tag{3.2.4}$$

$$= -\gamma_1 (w - w_i) + 4\imath f' \sum_j \Big[A^*_j A_{j+1} (u' + \imath v') . e^{-\imath\Delta t} . e^{+\imath(k_{j+1} - k_j)z} - A^*_{j+1} A_j (u' - \imath v') . e^{+\imath\Delta t} . e^{+\imath(k_j - k_{j+1})z}\Big] \tag{3.2.5}$$

$$= -\gamma_1 (w - w_i) + 4\imath u' f' \sum_j \left[A^*_j A_{j+1} . e^{-\imath\Delta t} . e^{+\imath(k_{j+1} - k_j)z} - c.c.\right] - 4v' f' \sum_j \left[A^*_j A_{j+1} . e^{-\imath\Delta t} . e^{+\imath(k_{j+1} - k_j)z} + c.c.\right] \tag{3.2.6}$$

$$= -\gamma_1 (w - w_i) - 8u' f' \sum_j \mathbb{I}\mathrm{m}\left[A^*_j A_{j+1} . e^{-\imath\Delta t} . e^{+\imath(k_{j+1} - k_j)z}\right] - 8v' f' \sum_j \mathbb{R}\mathrm{e}\left[A^*_j A_{j+1} . e^{-\imath\Delta t} . e^{+\imath(k_{j+1} - k_j)z}\right] \tag{3.2.7}$$

Note the RWA's above (for both polarization and inversion equations) discard modulations at frequency of multiples of ω_b. Quite a lot of physics has been removed by these RWA approximations, although it is very reasonable except in the very wideband limit. For example, the effect of next-nearest neighbour field components acting on the transition have been ignored, as have all more distant field-field interactions. In the next-nearest neighbour case, the dropped terms would impose a rapid ω_b oscillation onto the polarization ρ_{12}, which would in turn tend to impose sidebands at $\pm\omega_b$ onto each field component. It is reasonable to ignore such sidebands in the narrowband limit studied by most users of a multi-field Raman theory; but, in principle one might extend a multi-field theory to include them by inventing a scheme to apply the sidebands to the field component they are (near) resonant with.

C. Fields (A_j)

Note that the field evolution equation already has a carrier of $\exp\left[-\imath\left(\omega_0 t - k_0 z\right)\right]$ factored out of it. Thus I use A' from $A = A' \exp\left[-\imath\left(\omega_0 t - k_0 z\right)\right]$, not A. Finally, I need to insert the GFEA few-cycle correction to the polarization term, because my ($j \neq 0$) sub-envelopes A_j have an $\imath j\omega_b t$ time dependence that cannot be neglected.

From eqns.(2.7.21, 2.7.11), I get

$$\partial_z A'(t) = \imath\left[1 + \frac{\imath\partial_t}{\omega_0}\right] \frac{\zeta\sigma\omega_0 \bar{\alpha}'_{12}}{2\epsilon_0 c_0} A'(t) X(t) \tag{3.3.1}$$

$$\partial_z \sum_j A_j \exp\left[-\imath\left(\omega'_j t - k'_j z\right)\right] = \imath\left[1 + \frac{\imath\partial_t}{\omega_0}\right] \frac{\zeta\sigma\omega_0 \bar{\alpha}'_{12}}{2\epsilon_0 c_0} \left[\rho'_{12} e^{+\imath\omega'_b t} + \rho'^*_{12} e^{-\imath\omega'_b t}\right] \times \sum_j A_j \exp\left[-\imath\left(\omega'_j t - k'_j z\right)\right] \tag{3.3.2}$$

$$(\text{match } \omega_j \text{ terms})\qquad \left[\imath k'_j A_j + \partial_z A_j\right] e^{-\imath j\omega_b t} = \imath\frac{\zeta\sigma\bar{\alpha}'_{12}}{2\epsilon_0 c_0} \left\{A_{j+1}\rho'_{12} \exp\left[+\imath(k'_{j+1} - k'_j)z - \imath\Delta t\right] + A_{j-1}\rho'^*_{12} \exp\left(+\imath(k'_{j-1} - k'_j)z + \imath\Delta t\right)\right\} \times \left[\omega_0 + \imath\partial_t\right] e^{-\imath j\omega_b t} \tag{3.3.3}$$

Then, using

$$[\omega_0 + \imath\partial_t]\, e^{-\imath j\omega_b t} \;\longrightarrow\; \omega_0 e^{-\imath j\omega_b t} - \imath^2 j\omega_b e^{-\imath j\omega_b t} \quad\longrightarrow\quad [\omega_0 + j\omega_b]\, e^{-\imath j\omega_b t} \quad\longrightarrow\quad \omega_j e^{-\imath j\omega_b t} \tag{3.3.4}$$

So

$$\begin{aligned}
\left[\imath k'_j A_j + \partial_z A_j\right] e^{-\imath j\omega_b t} &= \imath\frac{\zeta\sigma\bar{\alpha}'_{12}}{2\epsilon_0 c_0}\left\{A_{j+1}\rho'_{12}\exp\left[+\imath(k'_{j+1}-k'_j)z - \imath\Delta t\right]\right. \\
&\qquad \left. + A_{j-1}\rho'^{*}_{12}\exp\left(+\imath(k'_{j-1}-k'_j)z + \imath\Delta t\right)\right\} \quad\times\quad \omega_j e^{-\imath j\omega_b t} \qquad (3.3.5)\\
\imath k'_j A_j + \partial_z A_j &= \imath\frac{\zeta\sigma\omega_j\bar{\alpha}'_{12}}{4\epsilon_0 c_0} u\left[A_{j+1}\exp\left[+\imath(k'_{j+1}-k'_j)z - \imath\Delta t\right] + A_{j-1}\exp\left[+\imath(k'_{j-1}-k'_j)z + \imath\Delta t\right]\right] \\
&\quad - \frac{\zeta\sigma\omega_j\alpha_{12}}{4\epsilon_0 c_0} v\left[A_{j+1}\exp\left(+\imath(k'_{j+1}-k'_j)z - \imath\Delta t\right) - A_{j-1}\exp\left(+\imath(k'_{j-1}-k'_j)z + \imath\Delta t\right)\right] \qquad (3.3.6)\\
\partial_z A_j &= \frac{\zeta\sigma\omega_j\alpha_{12}}{4\epsilon_0 c_0}\left\{-[v-\imath u]\, A_{j+1}\exp\left(+\imath(k'_{j+1}-k'_j)z - \imath\Delta t\right) + [v+\imath u]\, A_{j-1}\exp\left(+\imath(k'_{j-1}-k'_j)z + \imath\Delta t\right)\right\} \\
&\quad - \imath\,(k_j - k_0)\, A_j \qquad (3.3.7)
\end{aligned}$$

This is in agreement with both HPB[11] barring the opposite sign on the RHS – similar agreement occurs with SMN[17] once I identify $q = (v + \imath u)$. Note that generally $\Delta = 0$, as it just controls a frame rotation for ρ_{12}.

Note that we can assume, quite reasonably, that the multi-field envelopes A_j will be better behaved than the single-field envelope A. However, we have made *more* approximations, notably by RWA'ing away all the off-resonant cross terms driving the atomic transition so a multi-field approach is not always better. These off-resonant terms are at $2\omega_0 \pm \omega_b$ – see just prior to the starting point above of eqns.(2.6.25, 2.6.26).

In photon variables, the above field propagation equation is (using $R = 4\hbar\sigma$, also note $\zeta = 2$ to conserve energy)

$$\begin{aligned}
\partial_z A_{p,j} &= \frac{\zeta}{4} R\omega_j f_p\left\{-[v-\imath u]\, A_{p,j+1}\exp\left(+\imath(k'_{j+1}-k'_j)z - \imath\Delta t\right) + [v+\imath u]\, A_{p,j-1}\exp\left(+\imath(k'_{j-1}-k'_j)z + \imath\Delta t\right)\right\} \\
&\quad - (k_j - k_0)\, A_{p,j} \qquad (3.3.8)
\end{aligned}$$

IV. COMPARISONS

See *"Wideband pulse propagation: single-field and multi-field approaches to Raman interactions"* by Kinsler and New [1].

V. SUMMARY

I describe how to model a multi-frequency field such as that seen in a wideband Raman generation experiment using a single field envelope rather than a set of envelopes, one at each Stokes or anti-Stokes frequency. This requires that the field be propagated taking into account wideband effects, as described by either the SEWA theory of Brabec and Krausz [18], or the more general GFEA of Kinsler and New [7].

The usefulness of this single-field approach is not restricted to the Raman interaction described in this paper. It would be equally valuable for a near-degenerate optical parametric oscillator, or indeed any system where any two or more field components contain spectra that start to overlap as the pump or probe pulses get shorter.

It is important to note that it will usually only be more efficient to use a single-field simulation if pump pulses are very short, and effects like the next-nearest neighbour field interactions, neglected in the multi-field theory, need to be included, or if the extra computational overhead is not inconvenient. This is because in a single-field simulation, a very fine time-resolution is necessary to model the polarization and field oscillations closely enough to get good numerical convergence. However, this situation improves when the Raman transition has a smaller frequency compared to the pump pulse frequencies. One useful side effect of the fine time resolution is that it naturally gives a wide spectral bandwidth, so that many Stokes and anti-Stokes lines are modeled quite naturally. Further, our single-field model could be invaluable in modeling a short pulse pump-probe experiment where the probe frequency does not match any of the Stokes or anti-Stokes spectral lines generated by the pump pulse(s). A multi-field simulation would then need arrays for both the pump and probe Raman 'ladders" of Stokes/anti-Stokes lines, and the role of next nearest neighbour interactions (ignored in the multi-field model) could well become more significant.

In summary, the advantages of our single-field approach are twofold. First, it includes more physics than the multi-field approach, even compared to a multi-field approach enhanced by adding GFEA corrections to the propagation of the field components. Secondly, it deals effortlessly with the complications of overlapping spectra in the multi-field case.

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